RADIOACTIVE CONTAMINATION OF CISTERN WATERS ALONG THE CROATIAN COAST OF THE ADRIATIC SEA BY ^{90}Sr

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Abstract—**Measurements of radioactive contamination of water samples from cisterns collecting rainwater containing fission products from roofs and other surfaces have been carried out along the Croatian coast of the Adriatic sea since 1968. An exponential decline of radioactivity followed the nuclear moratorium. After the nuclear accident at Chernobyl, higher levels of ^{137}Cs and ^{90}Sr were detected again, with cistern waters being the only environmental samples in Croatia in which elevated ^{90}Sr activities persisted for several years. For the pre-Chernobyl period, the observed mean residence time of ^{90}Sr in cistern waters, estimated to be 6.2 ± 1.9 y, was similar to that calculated for fallout. Contrary, for the post-Chernobyl time, observed ^{90}Sr mean residence time was calculated to be considerably shorter, reflecting the tropospheric mean residence time. The annual dose for the critical adult population received from ^{90}Sr and ^{137}Cs by drinking cistern water was estimated to be very small, in the 1990's less than few $\mu Sv\ y^{-1}$. Health Phys. 77(1):62–66; 1999**

Key words: ^{90}Sr; water; contamination, environmental; fallout

INTRODUCTION

CISTERNS ARE artificial storage tanks for rainfall that has been collected from a roof or some other catchment area. In the Adriatic region in Croatia special catchment areas were sometimes built on the slope of a hill and could be very large. In the medieval Croatian town of Dubrovnik the top of the city walls served as a catchment area. Such cisterns were capable of providing water for whole villages and were used until the 1960's when major plumbing installations along the Croatian coast were built and fresh water supplies started to replace cistern water. However, even nowadays rain water from cisterns is sometimes used (on those places where fresh water is still unavailable, e.g., on some Adriatic islands) as a convenient source of drinking or technical water. Also, cistern water is frequently used for irrigation.

Although usually located underground, cistern reservoirs may be placed at ground level or on elevated stands, usually outdoors, but sometimes also within buildings. Cisterns are watertight and usually have smooth interior surfaces, enclosed lids, and are large enough to provide adequate storage. Therefore, they are excellent cumulative samplers of radioactive material introduced into the atmosphere by nuclear tests that were conducted in the atmosphere and by the release of the radioactive material from nuclear facilities.

This paper summarizes the measurements of ^{90}Sr and occasionally ^{137}Cs concentrations in cistern water samples along the Croatian coast of the Adriatic sea for the period 1968–1997, performed as a part of an extended monitoring program (Popović 1963–1978; Bauman et al. 1979–1992; Kovač et al. 1993–1998).

MATERIALS AND METHODS

Samples of cistern waters were taken once a year on nine regular locations along the Croatian coast of the Adriatic sea. Unfortunately, due to the great number of technical problems it was often impossible to collect samples from all investigated cisterns. There were nine sampling sites covering the Croatian coast of the Adriatic sea from Bale to Komaj.

Strontium, and occasionally ^{137}Cs, has been investigated since 1968. After the Chernobyl nuclear accident, ^{137}Cs has been investigated more regularly. Fallout samples were collected monthly in the town of Zadar.

Radiochemical methods were used to determine strontium levels. The radioactivity of ^{90}Sr was determined by beta-counting its decay product (^{90}Y) in a low-background, anti-coincidence, Geiger-Müller counter. Counting time was 80,000 s.

A gamma-ray spectrometry system based on a Ge(Li) detector (relative efficiency 15.4% with FWHM resolution of 1.87 keV at 1.33 MeV) coupled to a computerized data acquisition system (a 4,096-channel pulse height analyzer and personal computer) was used to determine ^{137}Cs levels in the samples from their gamma-ray spectra. The detector is shielded with 10-cm-thick lead that is lined with 2 mm of cadmium and 2 mm of copper. Samples were measured in Marinelli beakers, which were placed directly on the detector.

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Counting time for ^{90}Sr and ^{137}Cs measurements depended on sample activity but was never less than 60,000 s and was typically 80,000 s.

An efficiency calibration was carried out using sources provided by the International Atomic Energy Agency (IAEA) and World Health Organization (WHO).

Quality assurance and intercalibration of radioactivity measurements were performed through participation in the IAEA and WHO quality control programs.

RESULTS AND DISCUSSION

^{90}Sr activity concentrations

Nuclear tests conducted in the atmosphere and release of radioactive material from nuclear facilities cause the radioactive contamination of human environment. The fallout resulting from atmospheric dispersion of both short and long-lived radionuclides not only directly affects humans but also enters the food chain through the plants and animals and water supplies. ^{90}Sr and ^{137}Cs have been regarded as the fission products of a great potential hazard, due to relatively long half-life and similarity in metabolic processes to calcium and potassium, respectively.

^{90}Sr activity concentrations in cistern waters are given in Table 1. The activity concentrations of the water samples taken at the same time at different cisterns vary considerably. It can be explained by very different rates of water usage from respective cisterns, which depends upon the needs of the owner. Also, some owners sometimes exchanged all the water in order to clean and properly maintain the cistern reservoir. Therefore, residual fallout ^{90}Sr in some cisterns persisted for longer periods of time leading to high variations of individual ^{90}Sr mean residence times in respective cisterns. The highest ^{90}Sr activity concentration was found in 1968 in the cistern in the village of Doli in the very southern region of the Adriatic, which was 302.7 Bq m^{-3}. In late 1990's ^{90}Sr activity concentrations in cisterns were around 2 Bq m^{-3} (Fig. 1).

Mean residence time of ^{90}Sr in cistern water

Mean ^{90}Sr activity concentrations in cistern waters for the 1968–1997 period are presented at Fig. 2. An exponential decline of radioactivity followed the nuclear moratorium up to the Chernobyl nuclear reactor accident in 1986. Exponential decrease after the Chernobyl accident has a different slope. Generally, the Chernobyl accident did not cause any significant increase in ^{90}Sr activity in most of the environmental samples in Croatia. Unlike the atmospheric testing of nuclear weapons, the radionuclides that originated from the Chernobyl accident were not released directly into the upper atmosphere. As the result of the release mechanism and the

Table 1. ^{90}Sr activity concentrations (Bq m^{-3}) in cistern water samples.

		Locations								
Year	Mean	Bale	Brseč	Punat	Barić Draga	Pag	Rovanjska	Brela	Doli	Komaj
1968	196.8 ± 76.6				124.0				302.7	163.9
1969	104.1 ± 41.3				89.9				62.2	160.2
1970	105.3 ± 54.0	180.6					56.4		78.8	
1971	125.7 ± 49.8	184.3					62.5	130.2		
1972	73.8 ± 21.8	43.3			85.3			92.9		
1973	55.5 ± 14.2				69.7			41.3		
1974	50.6 ± 4.8				55.5			52.2		44.0
1975	48.2 ± 1.4						46.8			49.6
1976	45.2 ± 16.2		61.4						29.0	
1977	30.5 ± 4.4		29.2				33.3		24.1	35.5
1978	53.0 ± 9.0		65.9		52.9		40.3			52.9
1979	28.6 ± 5.6		20.7		33.3				31.8	
1980	35.1 ± 5.0				28.1		37.7			39.6
1981	33.3 ± 9.2				20.4		39.6		40.0	
1982	29.1 ± 12.5		31.0				10.8	28.4	46.1	
1983	14.7 ± 2.1		12.2				14.7	17.3		
1984	13.7 ± 1.4				12.0		13.7	15.4		
1985	13.8 ± 1.9							11.9		15.6
1986	79.9 ± 41.5	97.4	20.8		134.0					67.3
1987	33.8 ± 12.0	41.0		13.0		42.0				39.0
1988	31.4 ± 14.9		43.0	47.0		40.0	16.0			11.0
1989	15.3 ± 10.1		8.7	27.0		5.0	28.0			7.6
1990	7.0 ± 1.9		9.9	5.5		4.7	6.5			8.3
1991	9.3 ± 4.1			9.1	10.5	2.5	14.7	4.6	10.3	13.7
1992	9.0 ± 2.1	6.8	8.5	12.0	10.0	5.2		10.0	10.2	
1993	7.8 ± 2.2			10.0		5.6				
1994	4.4 ± 1.0	5.6				4.4	3.2			
1995	2.9 ± 0.9	4.0				2.9	1.8			
1996	1.7 ± 0.4	2.1				1.3				
1997	2.3 ± 1.0	1.4				3.3				

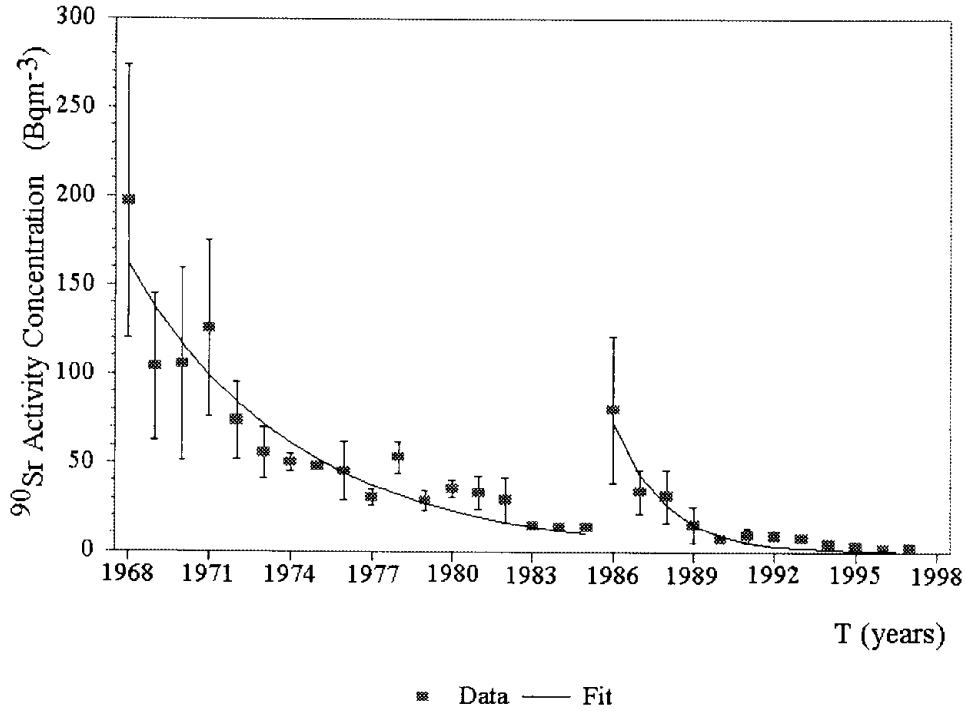

Fig. 1. ^{90}Sr activity concentrations in cistern waters for 1968–1997.

prevailing meteorological conditions at the time, the less volatile components of the Chernobyl debris (e.g., ^{90}Sr) were deposited closer to the accident location than the more volatile constituents (i.e., ^{137}Cs) (Aarkrog 1988; Franić and Bauman 1993). Thus, ^{90}Sr was only in minor quantities subjected to the global dispersion processes, as it was deposited to the Earth's surface within a period of a few days to a few weeks after the accident. In addition, changing meteorological conditions with wind from different directions at various altitudes and prolonged releases from a damaged reactor resulted in a very complex dispersion pattern over Europe. Consequently, the Adriatic region, except from the very northern part and very southern part, was initially unaffected by the plumes of contaminated air (UNSCEAR 1988; UNEP 1991). Also, the late spring and early summer of 1986 in Croatia were rather dry, leading to relatively low direct radioactive contamination, which was especially true for the Adriatic region (Bauman et al. 1979–1992).

However, due to the large catchment areas that are used to collect rain water, fallout radionuclides originated in Chernobyl were apparently "enriched" in cistern waters. Namely, dry fallout also entered the cistern reservoirs. Consequently, the cistern waters were the only environmental samples in Croatia in which ^{90}Sr, attributed to Chernobyl, persisted for several years after the accident. For comparison, in the city of Zadar a total of 90.7 Bq m^{-2} of ^{90}Sr was deposited in 1968. However, in 1986 (the year of the Chernobyl accident) only 7.7 Bq m^{-2} of ^{90}Sr were deposited compared to 6.6 and 6.2 Bq m^{-2} in 1985 and 1987, respectively. It should be noted that fallout has been collected only in the town of Zadar, in the mid-Adriatic region, that had been "missed" by the Chernobyl plumes. Therefore, low ^{90}Sr activity concentration in Zadar (44° 06′ N, 15° 13′ E) fallout does not necessarily reflect the ^{90}Sr activity concentration in cistern waters, especially in the year after the Chernobyl accident. For comparison, fallout in the Croatian capital of Zagreb (45° 48′ N, 16° 00′ E) in 1985, 1986, and 1987 was 6.9, 193.0, and 4.2 Bq m^{-2} of ^{90}Sr, respectively.

The mean data on ^{90}Sr activity concentrations in cistern water from Table 1 were fitted to the exponential function

$$A(t) = A(0)e^{-kt}, \tag{1}$$

where

$A(t)$ = ^{90}Sr activity concentration in cistern water at time t;
$A(0)$ = ^{90}Sr activity concentration in cistern water at zero time (years 1968 and 1986, respectively);
t = elapsed time; and
k = constant.

The reciprocal value of the constant k is the observed mean residence time of ^{90}Sr in the cistern water, i.e. $T_O = 1/k$. From eqn (1), T_O was found to be 6.2 and 1.9 y for 1968–1985 and 1986–1997, respectively. However, to find the real mean residence time, k should be corrected for radioactive decay. Therefore,

$$k = \lambda + k_R, \tag{2}$$

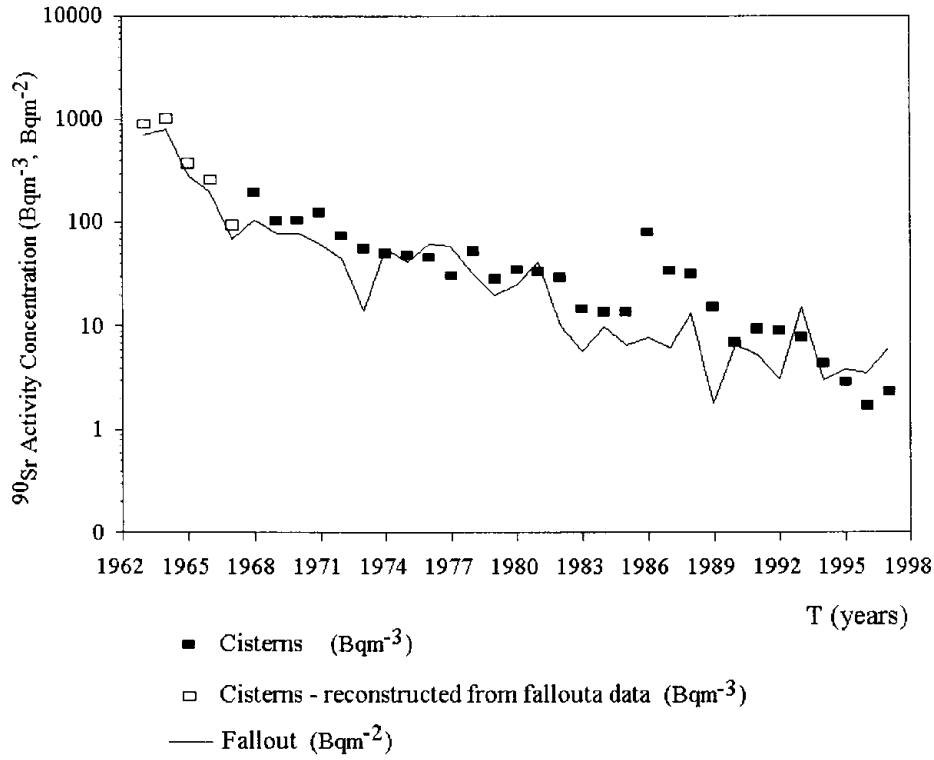

Fig. 2. ^{90}Sr activity concentrations in fallout and cistern waters for 1962–1997.

where $\ln(2)/\lambda = 29.12$ y is the half-life of ^{90}Sr.

From eqn (2) real mean residence time for ^{90}Sr in the cistern water, $T_M = 1/k_R$, was found to be 7.3 and 2.0 y for 1968–1985 and 1986–1997, respectively, which is just slightly higher than the observed mean residence times.

It should be noted that the observed mean residence time for the individual cisterns is considerably different, depending on the rate of water use. In order to obtain the standard deviations for respective periods, Monte Carlo simulations were performed. Uniform distribution has been assumed over the $A \pm \sigma$ value of ^{90}Sr activity concentrations shown in Table 1. For each year the random value was generated, and from such data was calculated the $1/k$ value using eqn (1). The process has been repeated 100 times. The mean value and standard deviation for $1/k$ were calculated to be 6.2 ± 1.1 and 1.9 ± 0.6 y for 1968–1985 and 1986–1997 respectively.

The ^{90}Sr mean residence time in cistern waters for the post-Chernobyl period reflects the tropospheric mean residence time, which is considerably shorter than the stratospheric mean residence time. Namely, the radioactive material from the damaged Chernobyl reactor did not reach the stratosphere in significant quantities, which is especially true for the less volatile radionuclides. Therefore, the pre-Chernobyl and the post-Chernobyl mean residence times of ^{90}Sr in cistern waters in addition to the rate of water usage apparently also depend upon the mechanism by which strontium was released into the atmosphere.

^{137}Cs in cistern waters considerably varied from location to location. Therefore, correlation between ^{137}Cs in Zadar fallout and ^{137}Cs in cistern waters was poor. Contrary, correlation between fallout ^{90}Sr concentrations and mean ^{90}Sr concentrations in cistern waters has been found to be fairly good, with the coefficient of correlation being $r = 0.85$ with $P(t) < 0.001$ and 28 degrees of freedom. In addition, after the Chernobyl accident ^{137}Cs concentrations in cistern waters were up to an order of magnitude greater than ^{90}Sr activity.

Dosimetry

Due to good correlation between ^{90}Sr in fallout and cistern waters from the known data on fallout, ^{90}Sr cistern water activity concentrations can be modeled by the equation

$$A_{cw}(t) = 1.29 \times A_{fall} + 6.38, \qquad (3)$$

where

$A_{cw}(t)$ = ^{90}Sr activity concentration in cistern water at time t in Bq m^{-3};
A_{fall} = ^{90}Sr activity concentration deposited on the Earth surface by fallout in Bq m^{-2}; and
t = elapsed time.

Therefore, it was also possible to reconstruct the ^{90}Sr doses for the periods for which only fallout data are available. Fig. 2 shows ^{90}Sr fallout activity in Zadar,

mean ^{90}Sr activity concentration in cisterns, and reconstructed data for the 1963–1967 period calculated by eqn (3). The calculated value for ^{90}Sr activity concentration in cistern water in 1963 is 924 Bq m^{-3}. The doses that would have been received by a hypothetical adult member of the public did not exceed 9.44 μSv y^{-1} (1963). Post-Chernobyl equivalent doses were trivial (<0.82 μSv y^{-1}). In the calculations the consumption of 1 L of cistern water per day has been assumed, which is consistent with the water balance of 3 L d^{-1} for Reference Man (ICRP 1975).

The dose-conversion used for the calculations were 2.8×10^{-8} and 1.3×10^{-8} Sv Bq^{-1} for ^{90}Sr and ^{137}Cs, respectively (IAEA 1996). The doses, on the order of magnitude of μSv y^{-1}, pose no significant radiation risk.

CONCLUSION

Analysis of cistern water radioactivity is a valuable tool for assessment of environmental radioactive contamination, especially for the fission radionuclides. The observed mean residence time of ^{90}Sr in cistern waters was estimated to be 6.2 ± 1.9 and 1.9 ± 0.6 y for pre-Chernobyl and post-Chernobyl periods, respectively. For the Croatian Adriatic region, estimated doses due to consumption of cistern water are small. However, the radioactivity should be regularly monitored.

REFERENCES

Aarkrog, A. The radiological impact of the Chernobyl debris compared with that from nuclear weapons fallout. J. Environ. Radioact. 6:151–162; 1988.

Bauman, A.; Cesar, D.; Franić, Z.; Kovač, J.; Lokobauer, N.; Marović, G.; Maračić, M.; Novaković, M. Results of environmental radioactivity measurements in the Republic of Croatia 1978–1991. Zagreb, Croatia: Institute for Medical Research and Occupational Health; Summary reports1979–1992 (in Croatian).

Franić, Z.; Bauman, A. Activity of ^{90}Sr and ^{137}Cs in the Adriatic Sea. Health Phys. 64:162–169; 1993.

International Atomic Energy Agency. International basic safety standards for protection against ionizing radiation and for the safety of radiation sources. Vienna: IAEA; 1996.

International Commission on Radiological Protection. Report of the Task Group on Reference Man. Oxford: Pergamon Press; ICRP Publication 23; 1975.

Kovač, J.; Cesar, D.; Franić, Z.; Lokobauer, N.; Marović, G.; Maračić, M. Results of environmental radioactivity measurements in the Republic of Croatia 1992–1997. Zagreb, Croatia: Institute for Medical Research and Occupational Health; Summary reports1993–1998 (in Croatian).

Popović, V. Environmental radioactivity in Yugoslavia 1962–1977. Belgrade: Federal Committee for Labour, Health and Social Welfare; Summary reports 1963–1978 (in Croatian).

United Nations Environment Programme. Mediterranean action plan, Assessment of the state of pollution in the Mediterranean Sea by radioactive substances. Athens: UNEP; 1991.

United Nations Scientific Committee on the Effects of Atomic Radiation. Sources, effects and risks of ionizing radiation. New York: United Nations; 1988.